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ANOMALOUS ^{14}C ENRICHMENTS IN THE EASTERN UK COASTAL ENVIRONMENT

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ABSTRACT. During the period from 1995 to 2011, radiocarbon measurements from the coast around Hartlepool in NE England have revealed anomalous enrichments in seawater, sediment, and marine biota. These cannot be explained on the basis of atomic weapons testing or authorized nuclear industry discharges, including those from the nearby advanced gas-cooled reactor. Enhanced ^{14}C-specific activities have also been observed since 2005 in biota during routine monitoring at Hartlepool by the Food Standards Agency, but are reported as "likely" originating from a "nearby non-nuclear source." Studies undertaken in Hartlepool and Teesmouth during 2005 and 2011 suggest that the ^{14}C discharges are in the vicinity of Greatham Creek, with activity levels in biota analogous to those measured at Sellafield, which discharges TBq activities of ^{14}C per annum. However, if the discharges are into Greatham Creek or even the River Tees, it is proposed that they would be much smaller than those at Sellafield and the high specific activities would be due to much smaller dilution factors. The discharge form of the ^{14}C remains unclear. The activity patterns in biota are similar to those at Sellafield, suggesting that initial inputs are dissolved inorganic carbon (DI^{14}C). However, the mussel/seaweed ratios are more akin to those found around Amersham International, Cardiff, which is known to discharge ^{14}C in an organic form. ^{14}C analysis of a sediment core from Seal Sands demonstrated excess ^{14}C to the base of the core (43–44 cm). ^{210}Pb dating of the core (0–32 cm) produced an accumulation rate of 0.7 g cm^{-2} yr^{-1}, implying that ^{14}C discharges have occurred from the 1960s until the present day.

INTRODUCTION

In 1995, the Management Committee of the UK nuclear industry commissioned SUERC to survey radiocarbon activities in the UK coastal marine environment (Cook et al. 1998). The purpose of this investigation was to determine the background ^{14}C level (from natural production and weapons testing fallout) in marine biota (mussels, crabs, flatfish, and roundfish) and the dissolved inorganic carbon (DIC) component of seawater, to determine accurately the influence of ^{14}C discharges from nuclear fuel cycle activities (predominantly from Sellafield, NW England) and other sources such as Amersham International (now part of GE Healthcare), Cardiff, south Wales. The majority of sampling sites were chosen with regard to their remoteness from nuclear sites, although four were located in the NE Irish Sea and North Channel to assess dispersion of ^{14}C from Sellafield (Figure 1). The UK marine background for 1995 was subsequently defined as 247.6 ± 1.0 Bq kg^{-1} C for DIC and marine biota (based on values from Burtonport, NW Ireland) and represents the value that should be subtracted from the activity derived for any samples to establish excess ^{14}C inputs for that year (Cook et al. 1998).

^{14}C discharges from the Sellafield nuclear fuel reprocessing plant, made predominantly as dissolved inorganic carbon (DI^{14}C) (Begg et al. 1992; Cook et al. 1995), dominated these studies, providing the largest point source of ^{14}C to the UK marine environment. The results indicated a significant ^{14}C enhancement in the DIC fraction of seawater close to Sellafield, but with substantial dilution with increasing distance, following the prevailing clockwise currents around the UK. Beyond the North Channel, excess DI^{14}C was relatively constant around the coast of Scotland to NE England. In SW England, DI^{14}C activities were indistinguishable from background. An equivalent data set was obtained for marine biota, again demonstrating significant excess ^{14}C in biota near to Sellafield, with sizeable reductions in specific activities beyond the North Channel. However, anomalously high ^{14}C-specific activities were noted for seaweed, roundfish, and crustaceans at Whitby, NE England, in comparison to samples collected from NE Scotland, suggesting that the enhancements were not Sellafield-derived. The advanced gas-cooled reactors (AGRs) at Torness (SE Scotland) and Hartlepool (NE England) (Figure 1) were identified as potential sources for this ^{14}C excess (Cook et al.

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1998); however, the only reported ^{14}C discharges made to coastal waters from these sites during this period was 22 MBq between 1993 and 1995 (MAFF 1994, 1995; RIFE 1996; Public Health England 2014). Discharges from Cap de la Hague in NW France and Amersham International were also discounted due to the low ^{14}C activities observed in samples from the SW of England site, which is in relatively close proximity to these potential sources.

Figure 1 Map of the UK and Ireland indicating sites referenced in the text.

Nevertheless, these anomalous results prompted further investigation in 1998/1999 (Gulliver et al. 2004) to determine the source of the ^{14}C enrichments. The results indicated significant ^{14}C enrichments in DIC and biota at Hartlepool, above ^{14}C levels measured either at Whitby (1995) or Torness (in 1999). Mussel/seaweed activity ratios of 10.2 (1998) and 18.3 (1999) were observed at Hartlepool, significantly higher than the equivalent ratio at Sellafield (2.0), which discharges ^{14}C as $DI^{14}C$. Seaweed readily utilize carbon from the DIC component of seawater (Lalli and Parsons 1993). This therefore suggested that the releases in the Hartlepool area were either sporadic as DIC, with biota integrating the ^{14}C over a period of time, or that the releases were similar to those at the Amersham International site, which produces labeled compounds employed in research, and diagnostic kits used in medicine for *in vitro* testing of clinical samples (MAFF 1995). This site is known to discharge ^{14}C in an organic form and a mussel/seaweed activity ratio of 10.0 was measured here (Cook et al. 1998). Gulliver et al. (2004) concluded that while it was possible that Sellafield ^{14}C discharges could be a contributor to DIC activities observed at Torness, they were not responsible for the DIC activities observed at Hartlepool.

In 2003, significant enrichments of ^{14}C in the high molecular weight dissolved organic carbon (DOC) fractions were noted by Ahad et al. (2006) in both the Tweed (~160 Bq kg^{-1} C excess) and Tyne estuaries (~80 Bq kg^{-1} C excess), situated 140 and 40 km north of Hartlepool, respectively, which are again unlikely to be Sellafield-derived.

In 2005, a small followup study of the Hartlepool area was undertaken from areas not previously

sampled. The results demonstrated significant ^{14}C enrichments and prompted a further, more detailed study, undertaken in 2011. The primary aims of this study were to (i) investigate the extent of ^{14}C enrichment in marine biota and sediments; (ii) identify, or constrain, the source(s) of ^{14}C enrichment and determine when this enrichment commenced; and (iii) provide an improved understanding of the speciation and transfer of ^{14}C in this coastal marine environment, which could form a decision support basis for the formulation and implementation of future monitoring programs.

In parallel with our research, the Food Standards Agency (see RIFE 1995–2014) undertook routine monitoring over the period 2005–2013 in the Hartlepool area and concluded that there were small enhancements in ^{14}C in seafood and, in particular, in mussel flesh at Hartlepool (RIFE 2006–2014). These ^{14}C enhancements were attributed ("in likelihood") to a nearby "non-nuclear source." Much of the land in the immediate vicinity of Seal Sands supports an industrial park for major chemical, pharmaceutical, energy, and petrochemical industries, as part of the North East England Process Industry Cluster complex (NEPIC), including Hartlepool AGR. In addition, the Greatham Creek area, lying to the west of Seal Sands, is bordered to the north and southwest by numerous industrial manufacturing and processing sites near Greatham and Billingham, respectively. During the timeframe of the studies discussed here, several industrial facilities at Billingham conducted operations involving the use and manufacture of radiochemicals, and were authorized to dispose of gaseous radioactive wastes including ^{14}C (RIFE 2000–2001). Furthermore, in 1999 sizeable ^{14}C enhancements were observed in grass samples taken at Billingham, although the source of this contamination was inconclusive (RIFE 2000–2001).

METHODS AND MATERIALS

In 2005, seaweed (*Fucus* sp.), mussels (*Mytilus edulis*), and limpets (*Patella vulgata*) were collected for ^{14}C analysis from three sites: Headland, Parton Rocks, and Greatham Creek (Sites 1–3, respectively; Figure 2). In 2011, biota [seaweed, mussels, winkles (*Littorina littorea*)] and a marsh plant (*Salicornia* sp.) were collected for ^{14}C analysis from four sites: Seal Sands north east (Site 4), Seal Sands south (Site 5), Greatham Creek (Site 6), and Headland (Site 7). Sediment cores were collected from Seal Sands (Sites 4 and 5) in addition to a surface sediment grab sample from Greatham Creek (Site 6) (Figure 2). Both 2005 and 2011 studies followed the same analytical procedures and are discussed together. Seal Sands is part of Teesmouth National Nature Reserve and written permission was obtained from the Senior Reserve Manager prior to sample collection.

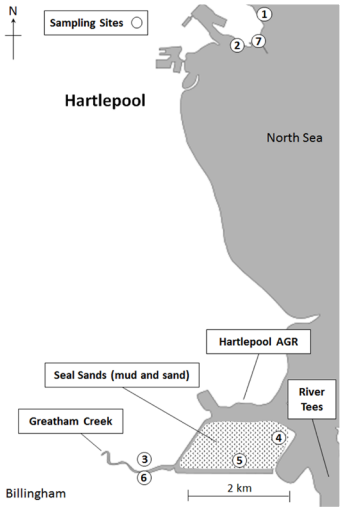

Figure 2 Hartlepool and Teesmouth area showing sampling sites visited in 2005 and 2011 (in circles).

Live mollusk samples, approximately 10–20 individuals of each species, were collected at each site. Soft tissue material was removed, washed thoroughly, freeze-dried, and homogenized to a fine powder. Shell material of the same live-collected specimens were retained for analysis and were washed and furnaced at 350°C to remove adhering organic matter. Shells were subjected to a 20% surface removal (by weight) using 1M HCl, dried, and homogenized to a fine powder. Seaweed and marsh plant samples (~500 g from each site) were collected, washed, dried at 50°C, and homogenized. Samples were combined (by species) for each site and subsampled for ^{14}C analysis (discussed below).

Sediment cores were collected from Sites 4 and 5 on Seal Sands using 12-cm-diameter (0.6-m-long) Perspex tubing, which was inserted into the relatively soft sediment by hand, with ~40 cm depth of sediment recovered in each case. Core 2 (Site 4) was selected for analysis by gamma spectrometry. The core was sectioned into 1-cm depth increments, oven-dried at 50°C, and ground into a fine powder. Organic ^{14}C analysis was undertaken on three horizons in Core 2, in addition to the grab sample from Site 6, and in each instance the samples were acid-washed with 1M HCl to remove inorganic carbonates, rinsed in deionized water, and oven-dried (40°C) prior to combustion and analysis by accelerator mass spectrometry (AMS). Samples undergoing gamma spectrometry were weighed (~20 g for each depth increment), hydraulically pressed into a uniform geometry, and then reweighed. Sediment samples were sealed in a polycarbonate container and stored to allow equilibration of ^{226}Ra daughters. High-resolution gamma spectrometry was performed on samples using an n-type high-purity germanium gamma (HPGe) detector. Detection efficiencies were calculated for ^{210}Pb (46.5 keV), ^{226}Ra proxies (at 295, 352, and 609 keV), and ^{137}Cs (661.6 keV) using in-house saltmarsh sediment standards prepared using certified tracer solutions.

Carbon dioxide was liberated from all samples for ^{14}C AMS analysis either by sealed quartz tube combustion (organic material) or by acid hydrolysis (shell carbonate), cryogenically purified with liquid N_2 under vacuum and converted to graphite according to the procedure of Slota et al. (1987). Sample $^{14}C/^{13}C$ isotope ratios were measured on the SUERC 250kV SSAMS (Freeman et al. 2008; Naysmith et al. 2010). Stable isotope ($\delta^{13}C$) ratios were measured on a VG SIRA 10 isotope ratio mass spectrometer. Results were calculated relative to the international standard (oxalic acid II, SRM-4990C) as ^{14}C activity ratios (fraction modern, F). Fraction modern results were converted to specific activities (Bq kg^{-1} C) using the regime for enhanced activity samples described by Mook and van der Plicht (1999).

RESULTS AND DISCUSSION

2005 Study

Results for samples collected in August 2005 are presented in Table 1. Much of the harbor area had undergone extensive redevelopment since the earlier SUERC studies and samples could not be collected from exactly the same locations as previously visited. ^{14}C-specific activities were lower in 2005 than in 1999 (Gulliver et al. 2004), but were still enriched relative to the natural/weapons testing background. The exception to this was seaweed at Site 3 (not previously sampled), which produced a ^{14}C-specific activity (3982.7 ± 14.0 Bq kg^{-1} C) that was significantly greater than the maximum observed in seaweed in 1995 near Sellafield (2352 ± 14 Bq kg^{-1} C), which discharges TBq quantities of ^{14}C per annum (Figure 3) (Cook et al. 1998). Unfortunately, no intertidal mollusks were available at Site 3 to determine the mussel/seaweed activity ratio. The ^{14}C-specific activities measured in seaweed at Sites 1 and 2 are 2 orders of magnitude lower than at Site 3, implying that these sites are somewhat remote from the source of discharges, potentially close to Site 3 (Figure 2).

Begg et al. (1992) observed activity patterns in biota near to Sellafield whereby the relative degree of ^{14}C enhancement was always mussels > winkles > seaweed, and proposed that the organism selectivity was related to the chemical form of ^{14}C discharge from Sellafield (DIC) and organism feeding behavior. This was confirmed by Cook et al. (2004) and showed, for a range of intertidal biota, that organism-specific feeding habits and trophic position strongly influenced ^{14}C uptake by biota, from an initial DI^{14}C source. A similar pattern emerges at Hartlepool, using limpet ^{14}C activities as a proxy for winkles (both graze intertidal algae and detritus) whereby the ^{14}C enhancement trend is mussels > limpets > seaweed. This indicates the possibility that ^{14}C uptake by biota at Hartlepool is from an inorganic ^{14}C source, similar in form to discharges observed at Sellafield. At this time, potential sources were identified as the large-scale petrochemical works or other local industry using ^{14}C as a tracer, local landfill/sewage receiving hospital or industry discharges, or a more remote facility around Billingham or Middlesbrough (with riverine transport).

Table 1 Excess ^{14}C-specific activities (Bq kg^{-1} C $\pm 1\sigma$) from the Hartlepool area (2005).

Site	Sample type	Excess ^{14}C-specific activity (Bq kg^{-1} C)
Headland (Site 1)	Seaweed	14.2 ± 1.3
Headland (Site 1)	Mussels	83.5 ± 1.5
Headland (Site 1)	Limpets	52.1 ± 1.3
Parton Rocks (Site 2)	Seaweed	3.9 ± 1.3
Parton Rocks (Site 2)	Limpets	28.9 ± 1.3
Greatham Creek (Site 3)	Seaweed	3982.7 ± 14.0

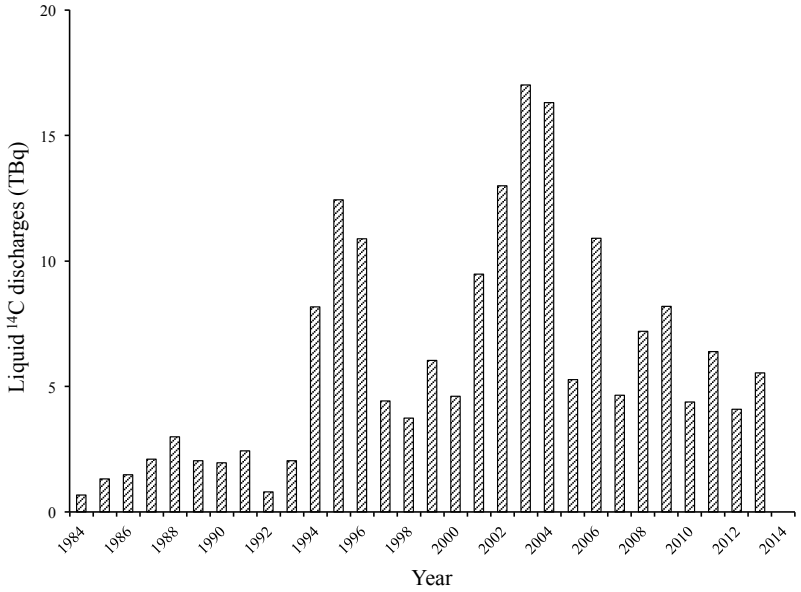

Figure 3 Annual Sellafield ^{14}C discharges (TBq) between 1984 and 2012

2011 Study

Following the 2005 study, a further investigation was undertaken in May 2011, focusing on the intertidal area at Greatham Creek and Seal Sands (an adjacent area of mudflats and sand) (Figure 2).

^{14}C results are presented for biota in Table 2 and sediment in Table 3.

Table 2 Excess ^{14}C-specific activities (Bq kg^{-1} C ±1σ) from the Hartlepool area (2011).

	Excess ^{14}C-specific activity (Bq kg^{-1} C)					
Site	Mussel tissue	Mussel shell	Winkle tissue	Winkle shell	Seaweed *Fucus* sp.	Marsh plant *Salicornia* sp.
Seal Sands: NE (Site 4)	945.2 ± 4.3	1485.1 ± 5.9	475.2 ± 2.8	896.6 ± 4.1	49.1 ± 1.5	11.1 ± 1.3*
Seal Sands: S. (Site 5)	1321.7 ± 5.9	2021.3 ± 7.4	696.7 ± 3.4	1870.3 ± 6.9	64.7 ± 1.4	—
Greatham Creek (Site 6)	—	—	1891.2 ± 5.2	2107.2 ± 6.4	175.6 ± 1.4	—
Headland (Site 7)	—	—	32.9 ± 1.5	28.6 ± 1.5	2.2 ± 1.2	—

*No ^{14}C background value had previously been determined for higher marsh plants (dicotyledons); therefore, values reported here are an approximation.

Table 3 Bulk organic sediment ^{14}C activities (Bq kg^{-1} C ±1σ) from selected sites (2011)

Site	Sediment fraction	Total bulk organic ^{14}C-specific activities (Bq kg^{-1} C ±1σ)
Greatham Creek (Site 6)	Surface grab	4661.6 ± 13.0
Seal Sands: Core 2 (Site 4)	7–8 cm	2829.6 ± 9.6
Seal Sands: Core 2 (Site 4)	29–30 cm	580.0 ± 2.2
Seal Sands: Core 2 (Site 4)	43–44 cm	299.7 ± 1.1

Biota

Substantial ^{14}C enrichments were observed in all biota samples (Table 2), analogous to those measured in biota close to Sellafield (Cook et al. 1998). This suggests either sizeable discharges from a remote location or that more modest discharges are occurring nearby but with less dilution. Variability in ^{14}C enhancements within biota observed between the closely neighboring Sites 4, 5, and 6 suggests that a smaller, localized ^{14}C input is more likely. ^{14}C activities follow a pattern whereby Greatham Creek > Seal Sands (S.) > Seal Sands (NE) > Headland. Biota tissue and seaweed show a threefold ^{14}C enhancement at Greatham Creek over winkle tissue and seaweed at Seal Sands, suggesting a ^{14}C source exiting Greatham Creek prior to seaward dispersion to Seal Sands. At Seal Sands, the relative degree of ^{14}C enhancement is mussels > winkles > seaweed, similar to ^{14}C results from 2005 and to Sellafield studies (Begg et al. 1992; Cook et al. 2004), alluding to initial inorganic DI^{14}C discharges.

Most shell carbonate is formed from the DIC in surrounding water (McConnaughey et al. 1997; Gilliken et al. 2006) and metabolic processes typically contribute only ~10% to shell carbonate, although this can vary by up to 25–40% (atypically) (Gilliken et al. 2005, 2006, 2007) depending on species, age, and environmental conditions. The results here indicate that mussel shell ^{14}C-specific activities exceed corresponding tissue-specific activities by 36% and 35% at Sites 4 and 5. Winkle shells showed a greater difference, at 47% and 63% for Sites 4 and 5, respectively. The trend in winkles at Greatham Creek was less pronounced (10%) and was slightly reversed in biota tissue at Site 7 where ^{14}C enhancements over background were relatively small. Again, the higher activities in the shell compared to soft tissue would seem to imply a discharge in the inorganic form.

Mussel to seaweed specific activity ratios of 19.2 and 20.4 were observed at Sites 4 and 5, respectively, and, in contrast to the above, this could imply an organic ^{14}C form of discharge. Low ^{14}C-specific activities in seaweed support this assertion, as these species will readily utilize available

DIC from water. Therefore, higher comparable ^{14}C activities in seaweed would be expected if the discharge was in the inorganic form. However, this is contrary to the enhanced ^{14}C-specific activities observed in *Fucus* sp. in 2005 and activity trends in both studies that imply that a significant DI^{14}C discharge had been made in the area.

Sediments

Selected sediments were analyzed for bulk organic ^{14}C-specific activities, and the results are presented in Table 3. Substantial ^{14}C enrichment is apparent in the surface grab from Greatham Creek, supporting the suggestion that the ^{14}C source is in this vicinity. Site 4 Core 2 comprises sandy mud to a depth of 30 cm, at which point there is an abrupt change in composition to sand. Excess ^{14}C is apparent in all depth increments that were analyzed and shows a trend of decreasing activity with depth. Gamma spectrometry was also performed on Core 2 in an attempt to define a temporal framework for the ^{14}C contamination using the constant flux: constant sedimentation (CF:CS) excess ^{210}Pb dating model (Appleby and Oldfield 1992), alongside a potentially supporting ^{137}Cs profile. The results are presented in Figure 4. The excess ^{210}Pb profile shows a trend of decreasing specific activity with depth from around 4–32 cm. A plot of the natural logarithm of these data versus cumulative mass is illustrated in Figure 5. From the linear equation derived from these data, an accumulation rate of 0.7 g cm^{-2} yr^{-1} was determined. This places the abrupt change in sediment composition from sandy mud to sand at around 1973, which coincides with the last extensive period of engineering undertaken at Seal Sands between 1971 and 1974, designed to establish sea walls and achieve land reclamation (Davidson and Evans 1986). The change in sediment composition may well have arisen in response to the engineering works reducing tidal energy/flow at Seal Sands, thereby increasing the propensity for fine-grained sediment to settle-out during periods of inundation. The presence of excess ^{14}C below this change in sediment composition strongly implies that discharges were occurring prior to the 1970s.

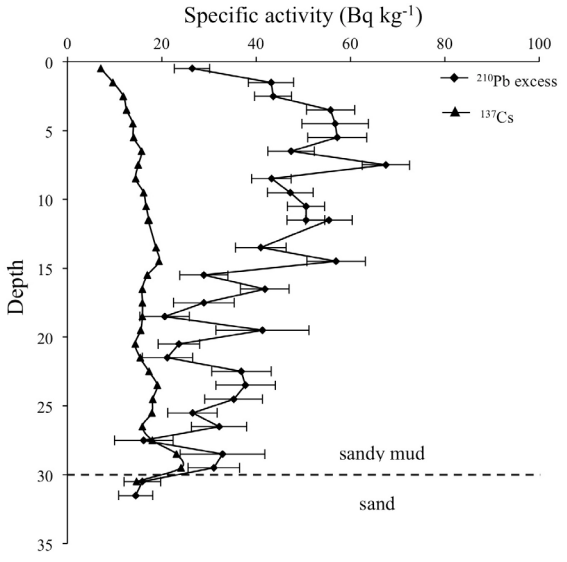

Figure 4 Core 2 (Site 4) sediment core profile showing ^{137}C and unsupported (excess) ^{210}Pb-specific activities (Bq kg^{-1}) and the sharp transition (dotted line) from sandy mud (above) to sand (below). ^{137}Cs uncertainties (<3% at ±1σ) omitted for clarity.

^{137}Cs is present throughout the core and there is a systematic decrease in the specific activity from 15 cm depth to the surface, indicative of the decreasing Sellafield discharges and redissolution of the general ^{137}Cs burden from sediment into the water column, and also demonstrating that the sediment

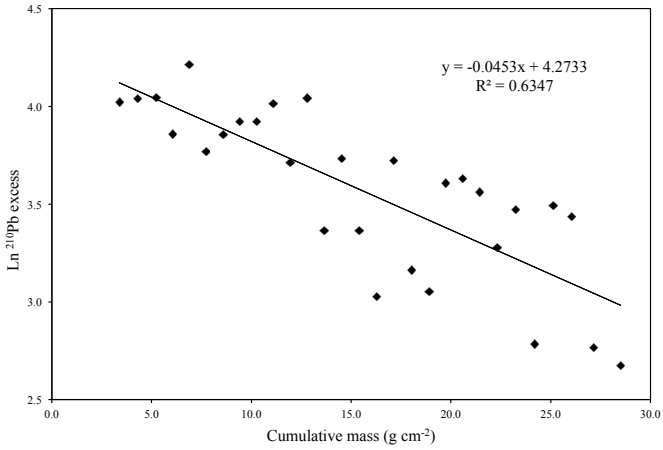

Figure 5 $Ln^{210}Pb_{excess}$ versus cumulative mass for Seal Sands Core 2.

is not well mixed. Three poorly defined ^{137}Cs peaks are apparent at 15, 24, and 30 cm, which could conceivably be attributed to the major inputs of ^{137}Cs from Chernobyl (1986), Sellafield (1960s to 1970s), and the fallout maximum (1963), but the excess ^{210}Pb chronology does not support this, attributing sediment deposition ages for these peaks to 1993, 1981, and 1974, respectively. However, these peaks do correspond to small peaks in the excess ^{210}Pb activity within the general trend of decreasing activity with increasing depth and could be indicative of transient changes in sediment composition. Plater and Appleby (2004) reported a ^{210}Pb chronology from the west side of Seal Sands that was limited by a poorly resolved ^{137}Cs profile. Therefore, it is possible that the sediment accreting in Seal Sands originates from a source where the ^{137}Cs is well mixed. They also observed rapid sediment accretion, especially over the 25 years prior to their study and attributed this to the engineering works and land reclamation activities.

CONCLUSIONS

Studies undertaken at SUERC over the period 1995–2011 demonstrate significant ^{14}C enrichments in intertidal biota and sediments in the Teesmouth area. The AGR at Hartlepool is an improbable source since there were no authorized liquid discharges from this facility during the period of investigation (RIFE 1995–2014; Public Health England 2014), other than a very minor discharge of 22 MBq in 1993–1995, and current ^{14}C discharges are made *via* the atmosphere. Routine monitoring by the Food Standards Agency has led to the postulation of a "nearby non-nuclear" source as the "likely" source of contaminant ^{14}C, although no further details have been published.

While ^{14}C-specific activities in biota are analogous to those observed near to Sellafield, which discharges TBq quantities of ^{14}C to the Irish Sea, patterns of ^{14}C dispersion in the Teesmouth area suggest that more modest discharges occur nearby, with significantly less dilution. In the case of Sellafield, discharges are directly into the NE Irish Sea. At Teesmouth, ^{14}C activity patterns and near-field heterogeneity in biota suggest that the source is most likely from the immediate vicinity of Greatham Creek and that this, or the nearby River Tees, is the conduit for supply of ^{14}C to Seal Sands and beyond.

The form of ^{14}C discharge remains unclear, although the trend in activities (mussels > winkles > seaweed) is the same as found at Sellafield that is known to discharge ^{14}C as DIC. However, in contrast, there is some evidence, in the form of the high mussel/seaweed activity ratio, to suggest that the initial input may have been organic. Using the accumulation rate established from the excess

^{210}Pb and the change in sediment composition proposed to have occurred in the early 1970s, it can be inferred that ^{14}C discharges preceded the engineering works.

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